



Pump-Probe Studies of K-Shell Photoionization and vacancy Decay of Alkali Atoms at the ALS

Ali Belkacem

Chemical Sciences Division

Funding: BES/DOE

Collaborators



Marc Hertlein

Hidehito Adaniya

Kyra Cole

Ben Feinberg

Jason Maddi

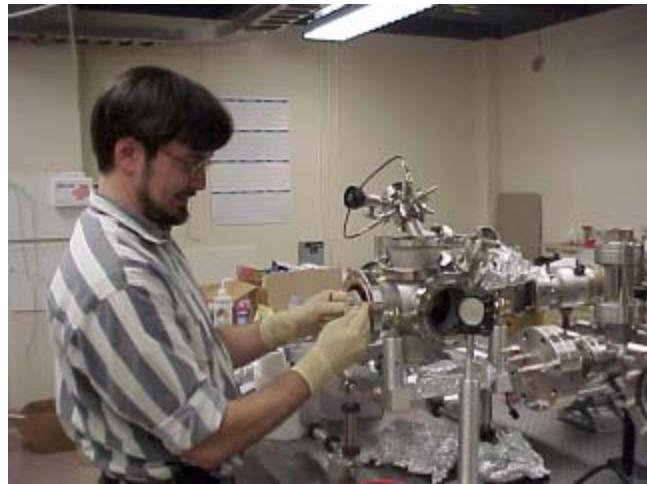
Nadine Neumann

Timur Osipov

Mike Prior

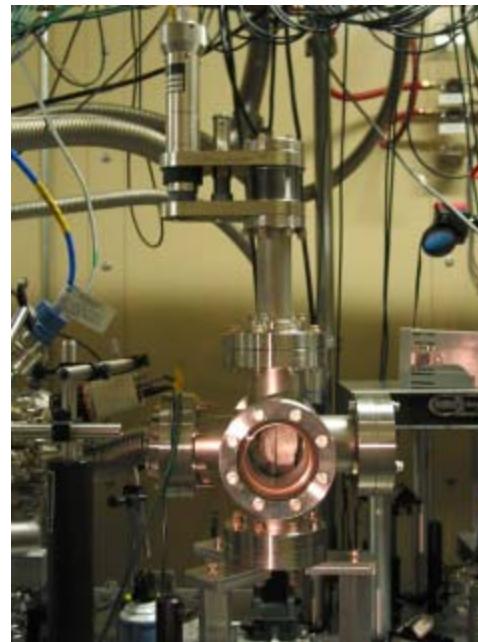
Ralf Schriel

Apparatus on beam line 5.3.1

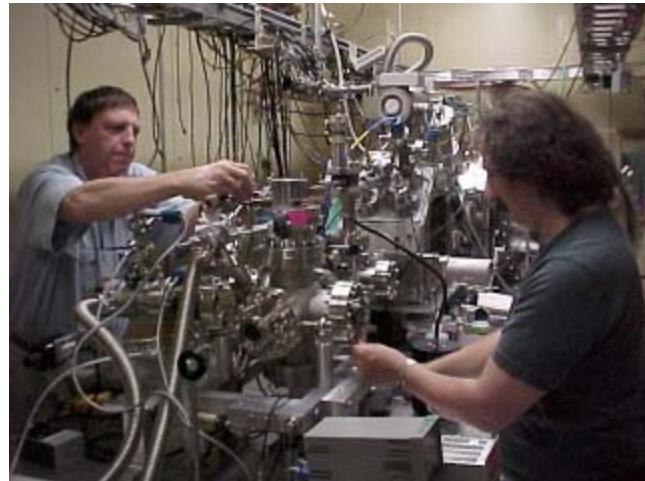


Marc

Merging laser mirror

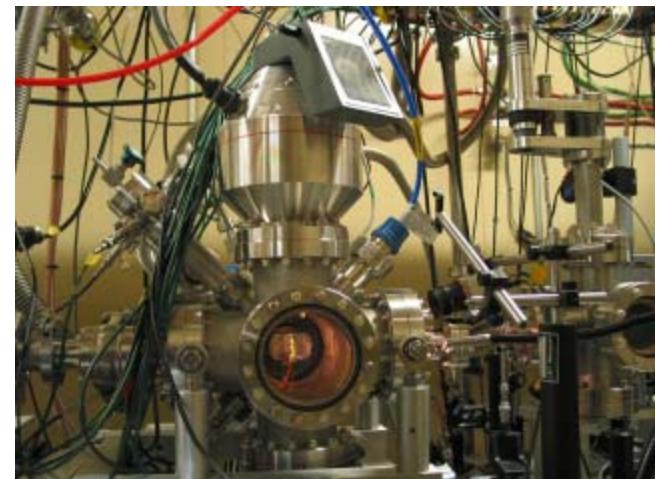


Jason and Ralf



Mike

Ali





Two aspects in these studies

Synchrotron x-rays are used as a probe of the structure of the laser dressed atom or molecule and the time evolution of the **laser-excited system**

Laser = pump

x-ray = probe

The relaxation of the “**x-ray-excited or ionized**” atom or molecule is modified by the presence of the high intensity laser field.

X-ray = pump

Laser = probe

Scope



Goals:

- Use of ultrafast *keV-energy* x-rays to study time-resolved inner-shell excitation and relaxation in atoms and molecules in gas phase.
- Study Auger dynamics at sub-femtosecond time scale

ps fs as

Methods:

- Development of techniques applicable from picosecond to attosecond time scales
- ultrafast laser pulses / electric fields to modify x-ray/target interactions
- exploiting fast electron correlation between inner + outer electrons
- pump-probe techniques, laser / electric field dressing



Approach

Gas phase targets



Direct target probe

- Directly probe target products of interaction
- Thin target - insensitive to x-ray/laser interaction length mismatch
- Zero-background type measurements possible
(Works well on paper)
- Large effects
- Techniques extendible to any gas target

Valence electrons

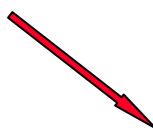
- respond to core dynamics
- can be excited/ionized during core decay
- significantly affect core electron binding energies



Core electrons

- interact with valence electrons after excitation / ionization (PCI effects, ...)
- respond to valence electron changes
- fast (fs) time scale dynamics

Correlation



Configuration interaction

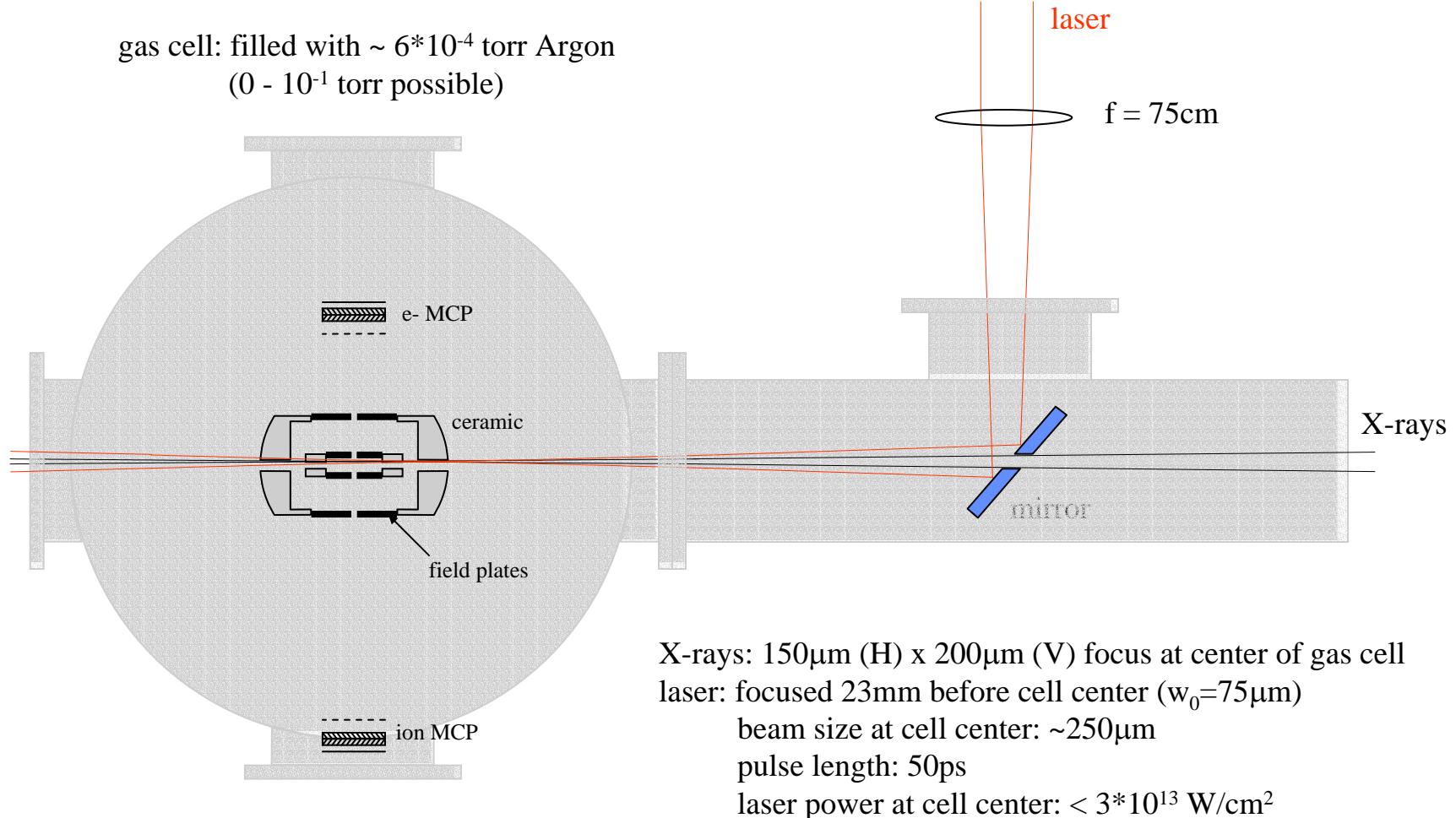
- Valence electrons can be used as a control handle on atom / X-ray interactions
- Core hole relaxation can change as a function of valence electron configuration
- Ion charge states produced by X-ray core excitation change as a result of valence electron configuration

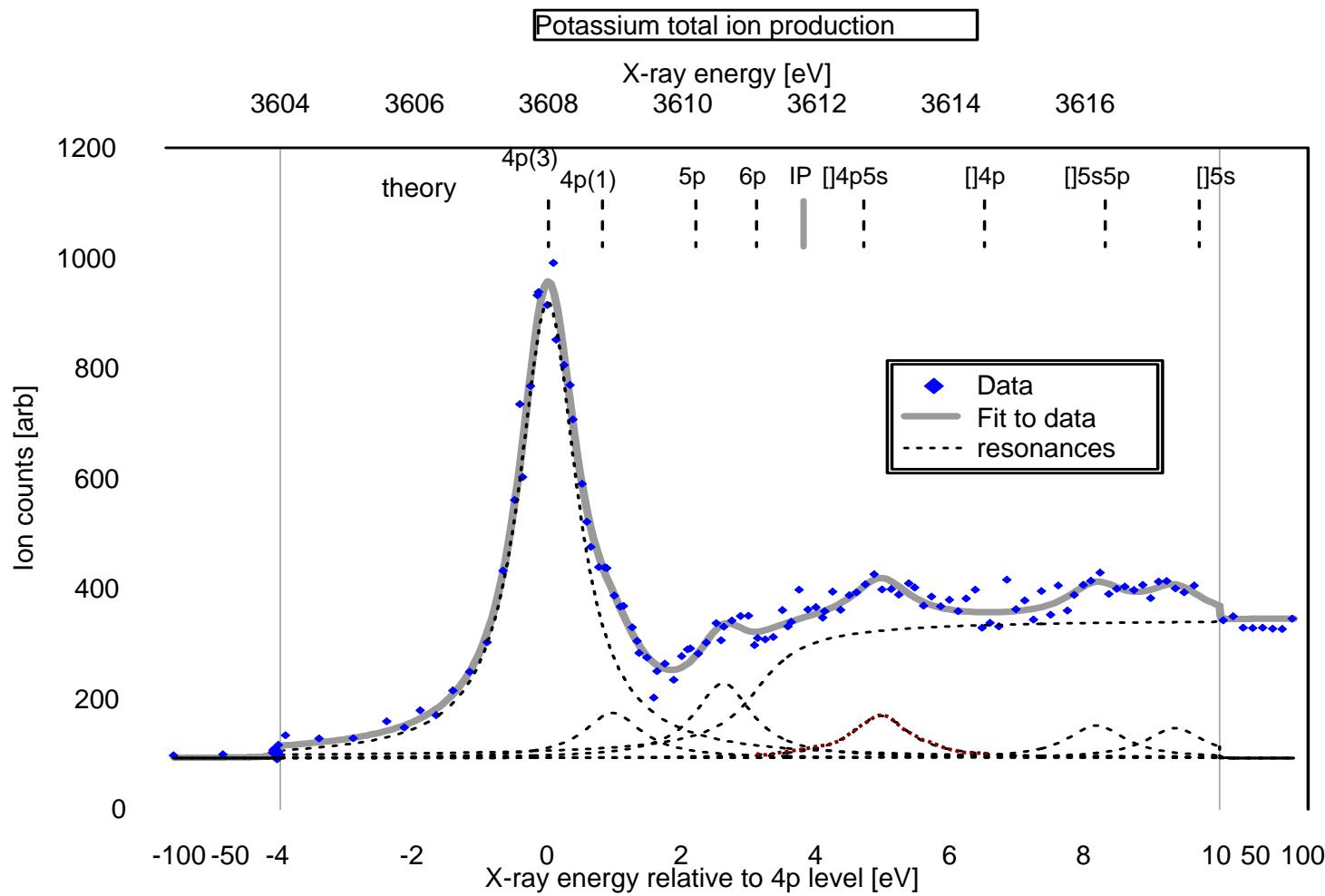
Experiment setup beam line 5.3.1

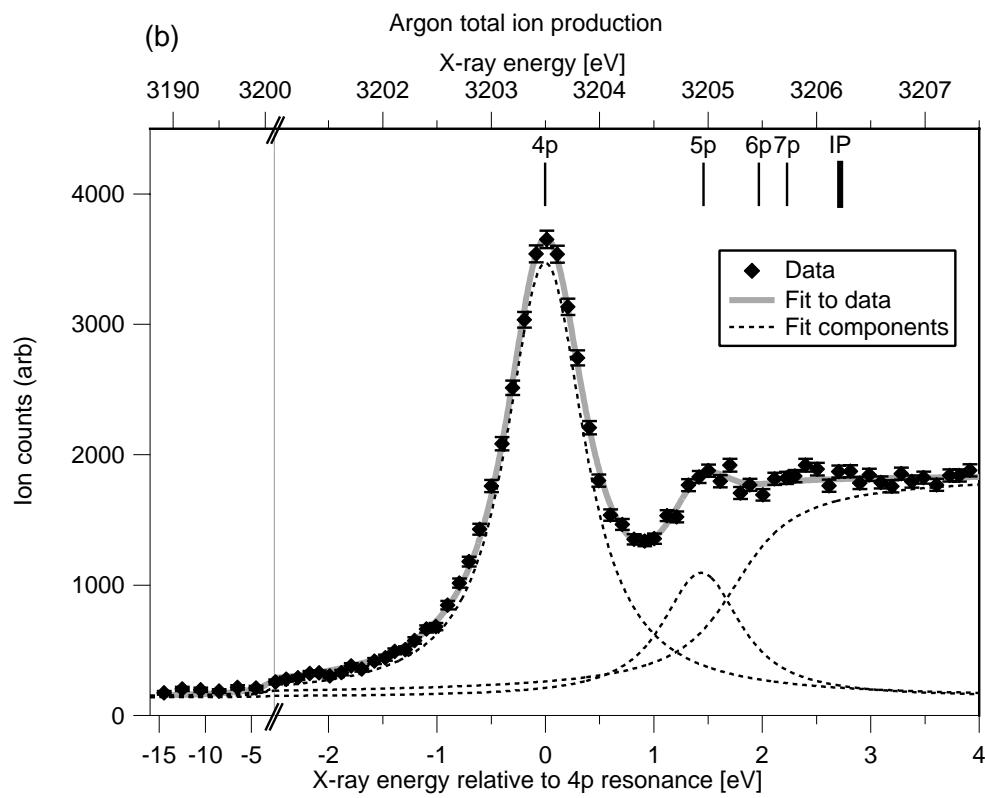


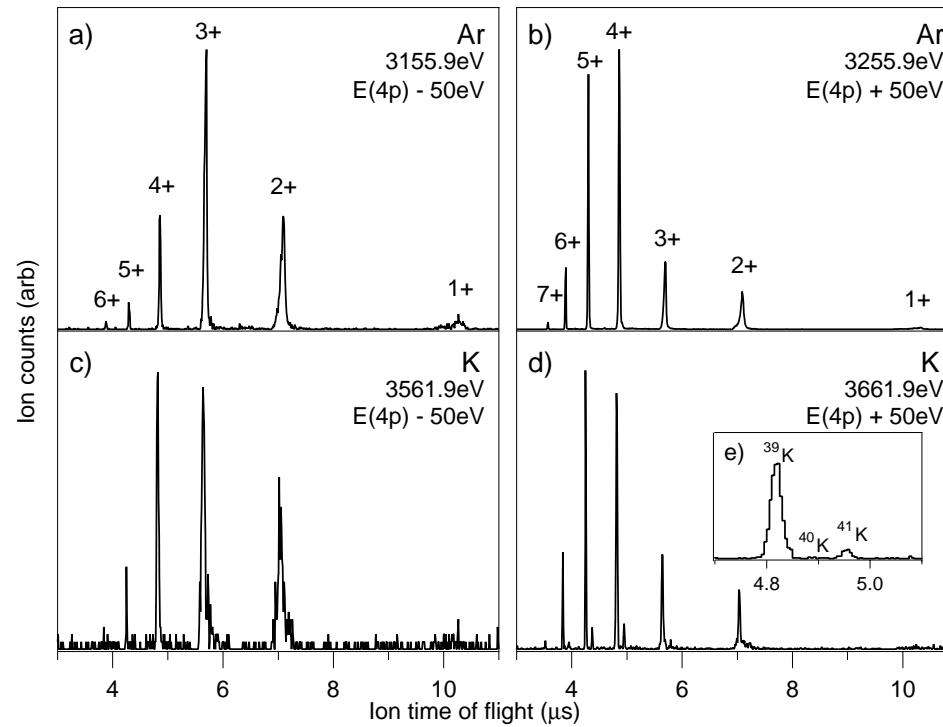
X-rays and laser pulses overlap in Argon gas cell

gas cell: filled with $\sim 6 \times 10^{-4}$ torr Argon
(0 - 10^{-1} torr possible)

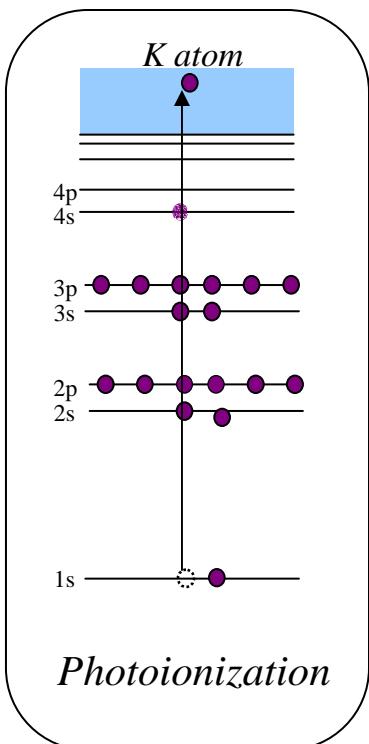




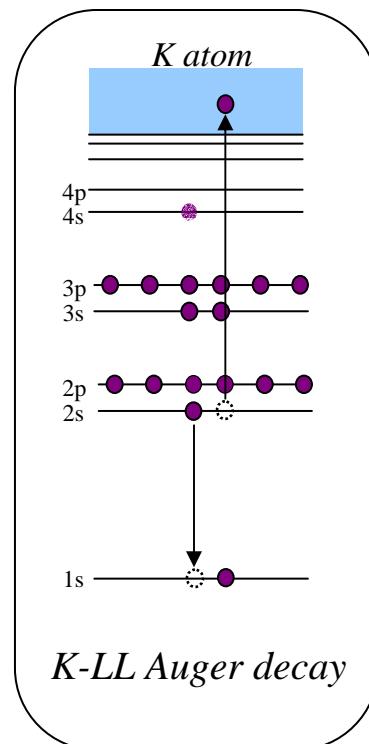




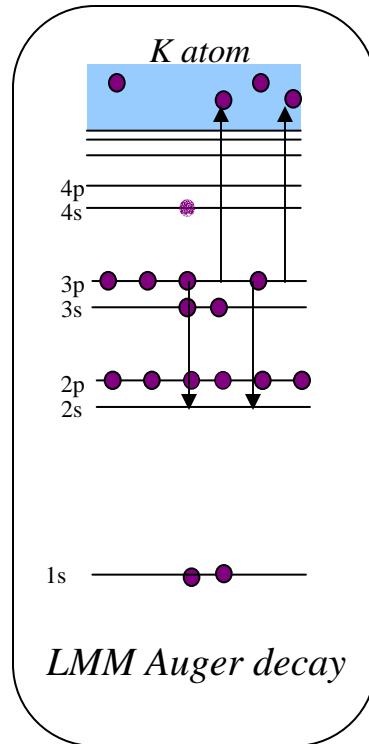
Photoionization

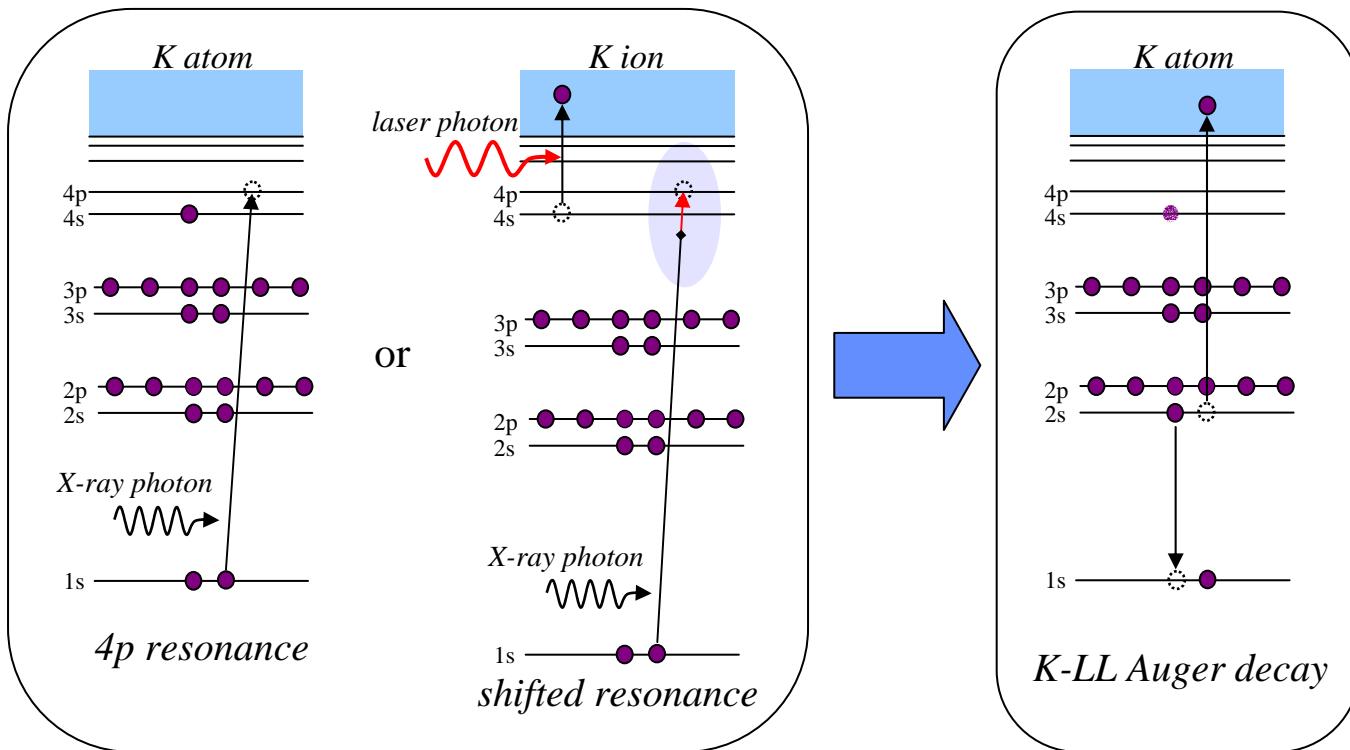


KLL Auger



LMM Auger and multiple ionization





Potassium



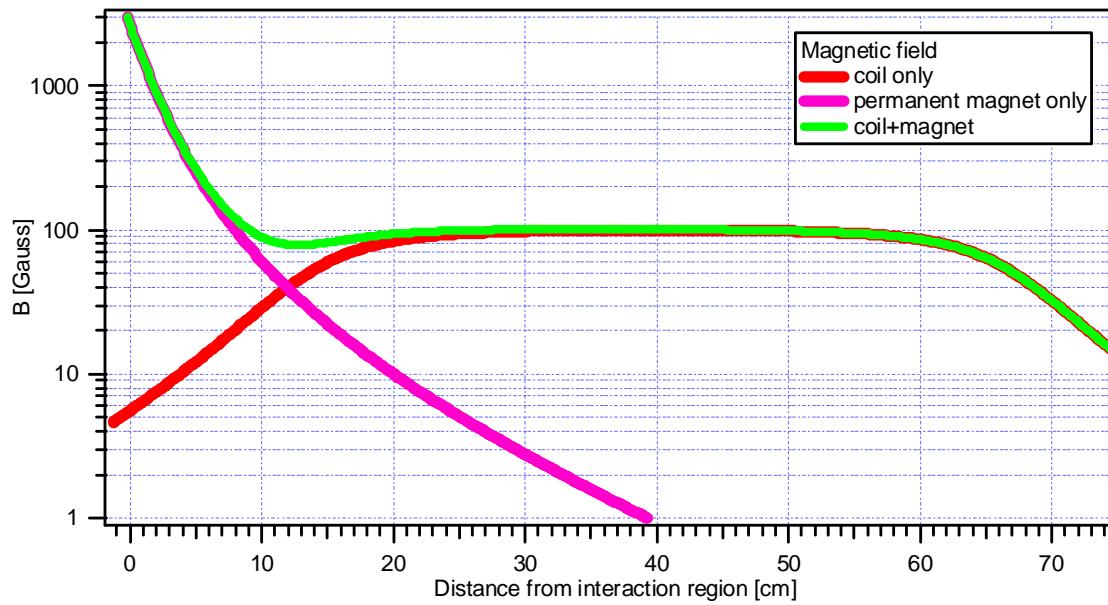
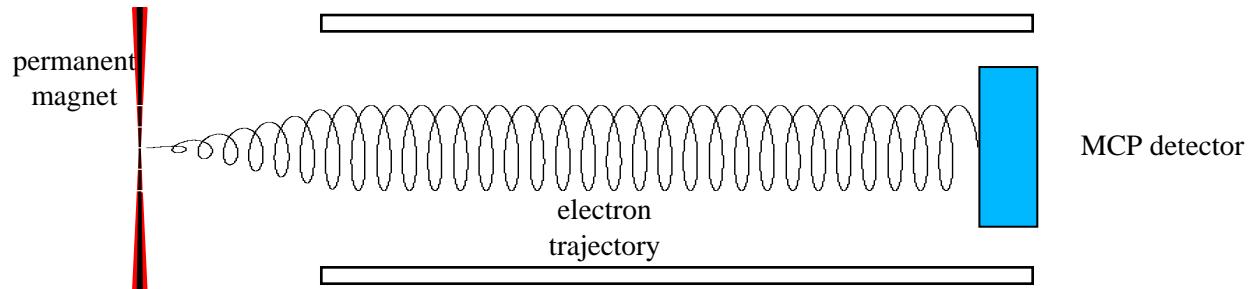
Good candidate for electron correlation studies:

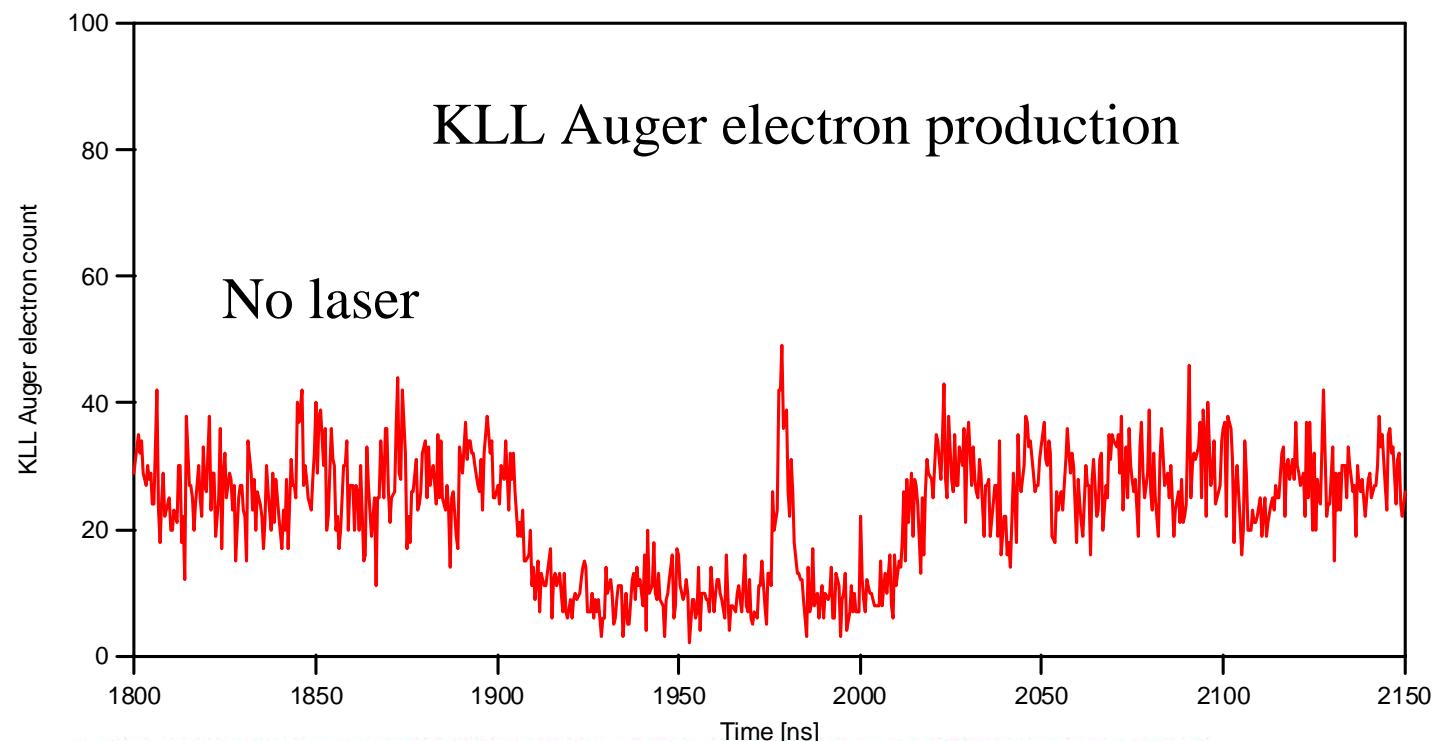
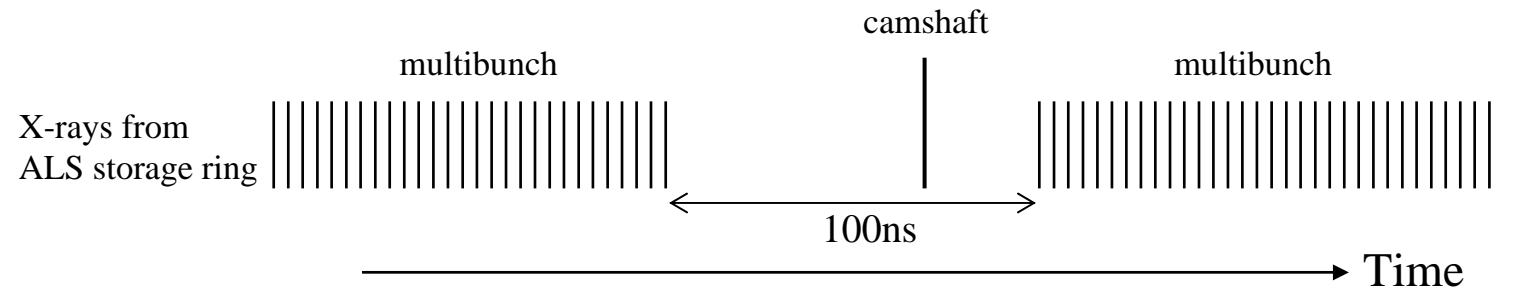
- Valence 4s electron has large probability of presence at the core and therefore influences 1s electron
- Outer electron is easily accessible by laser photons

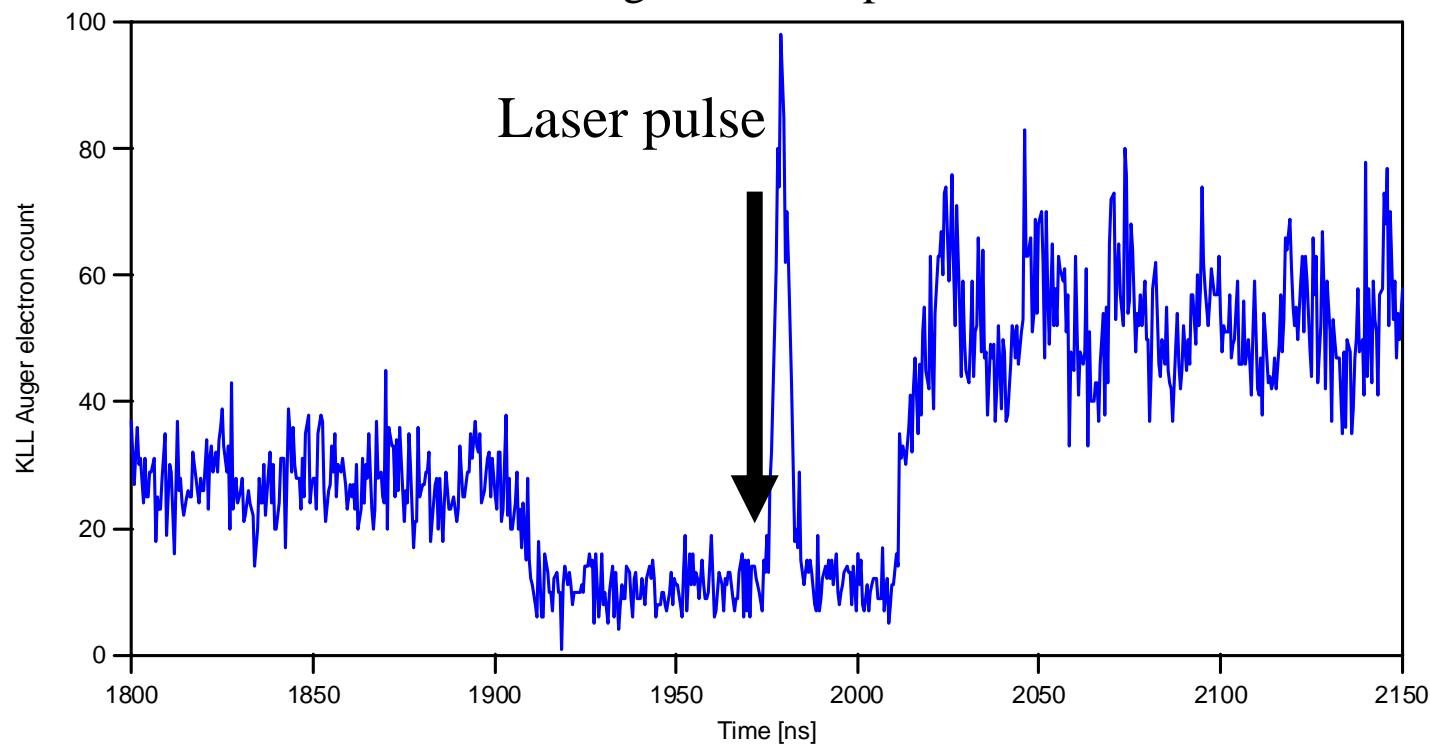
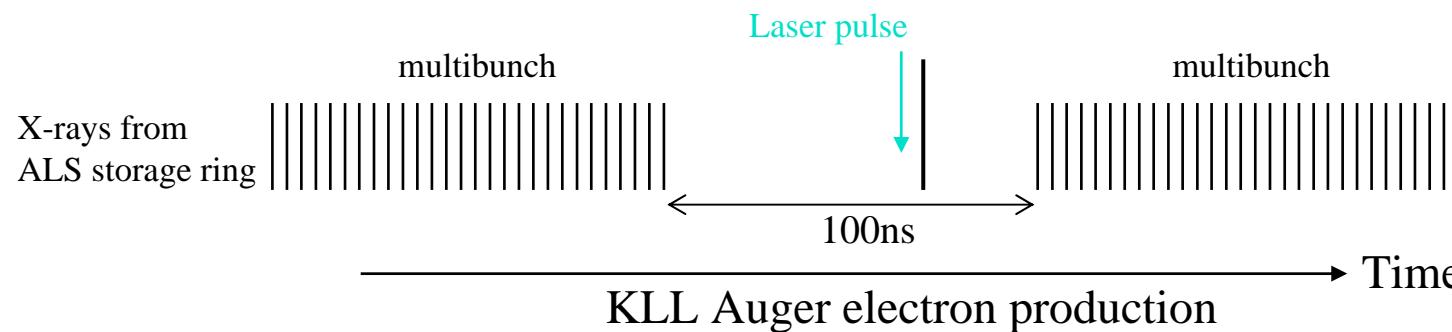
Flip side: Pain in the neck – creates large backgrounds when laser-on and loves to contaminate MCP detectors.

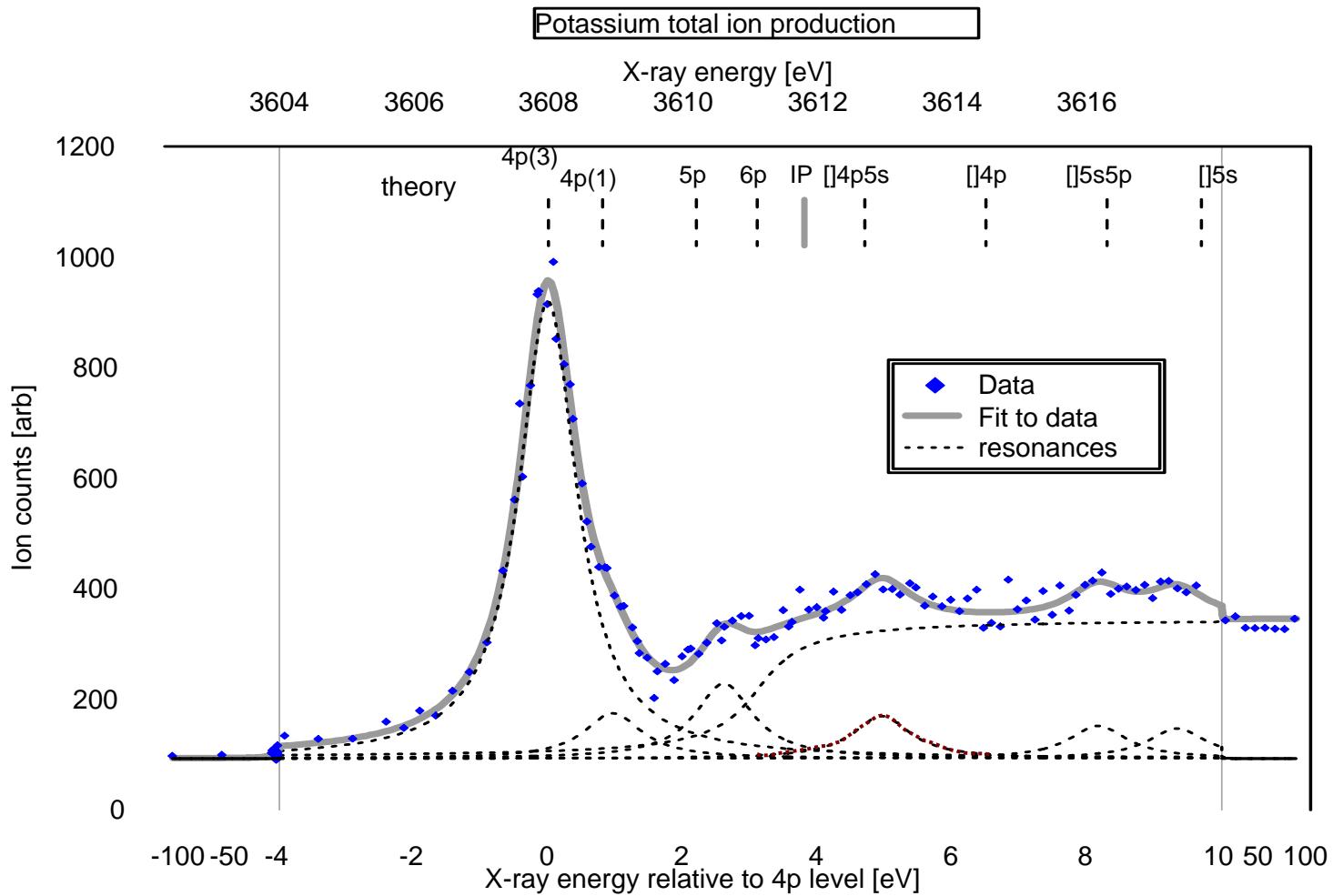
Experimental method:

- Only allow high energy (KLL Auger) electrons
Near zero background
- Use adiabatically decreasing strong magnetic field ($>3\text{kG}$) for high collection efficiency of energetic electrons
- Using tunable x-rays (3.6keV) from ALS synchrotron (LBNL) with collinear 100fs 800nm laser pulse

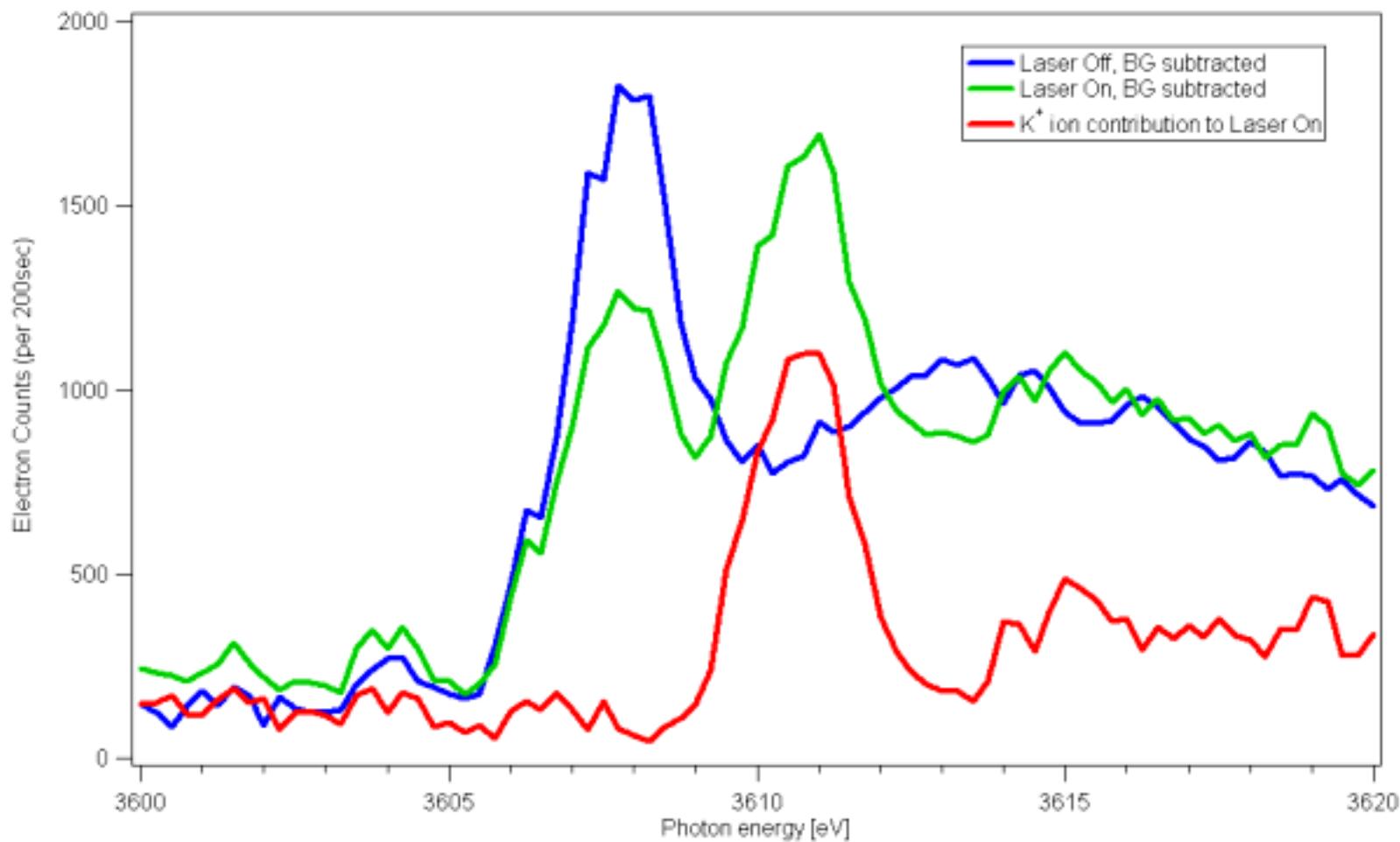


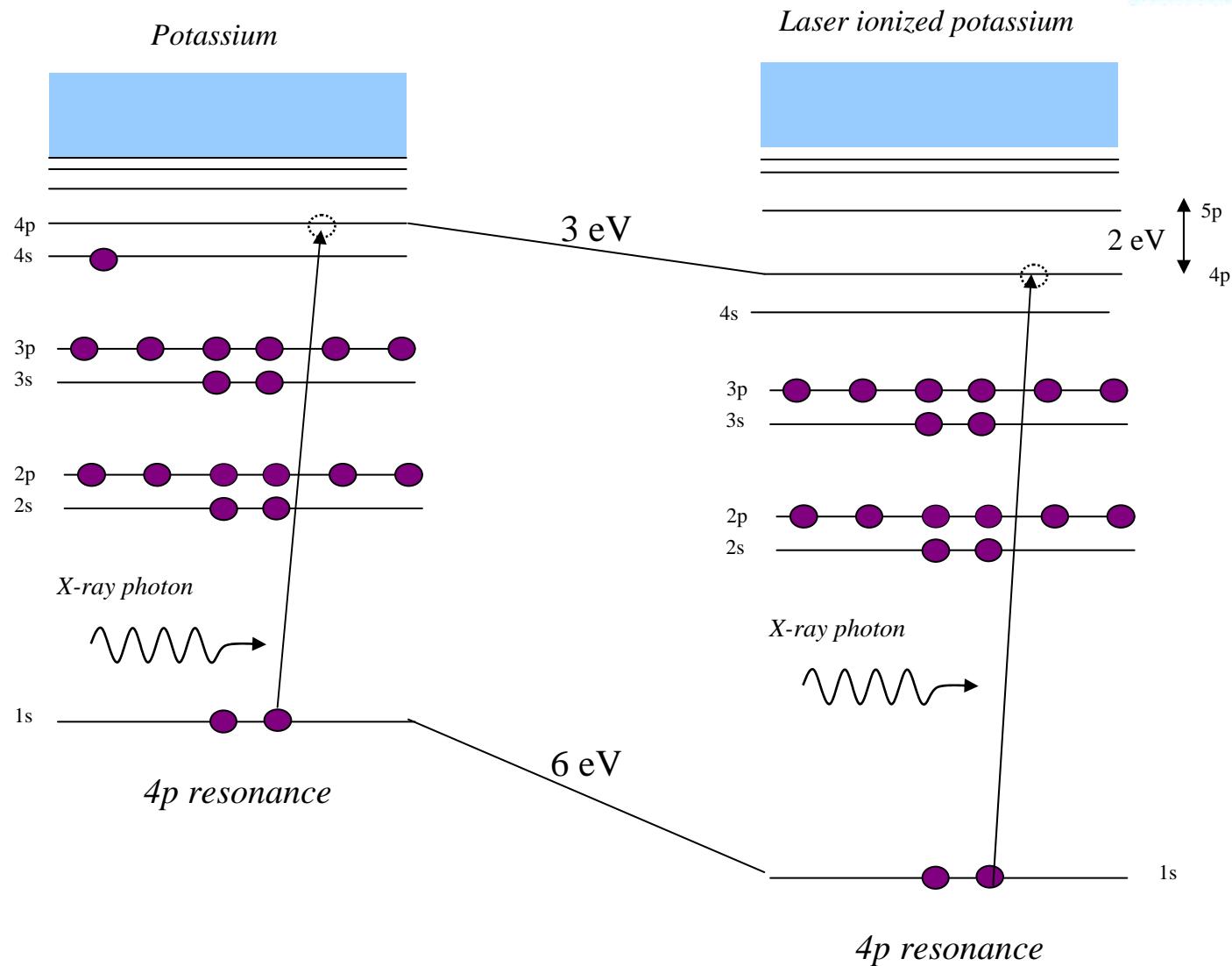


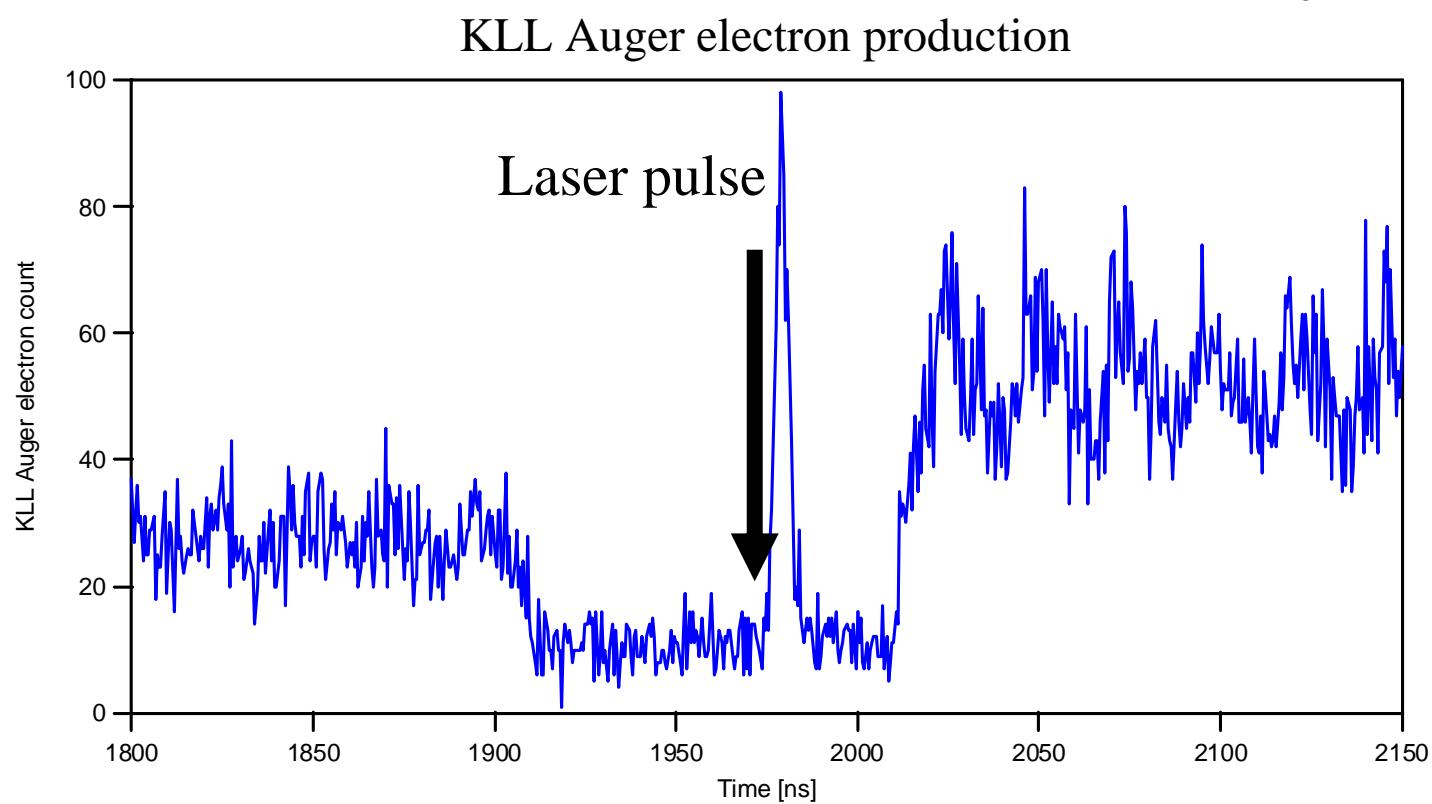
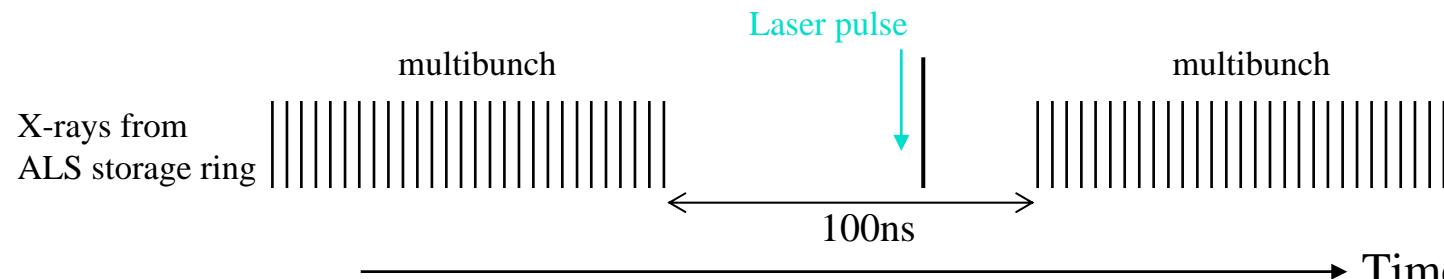




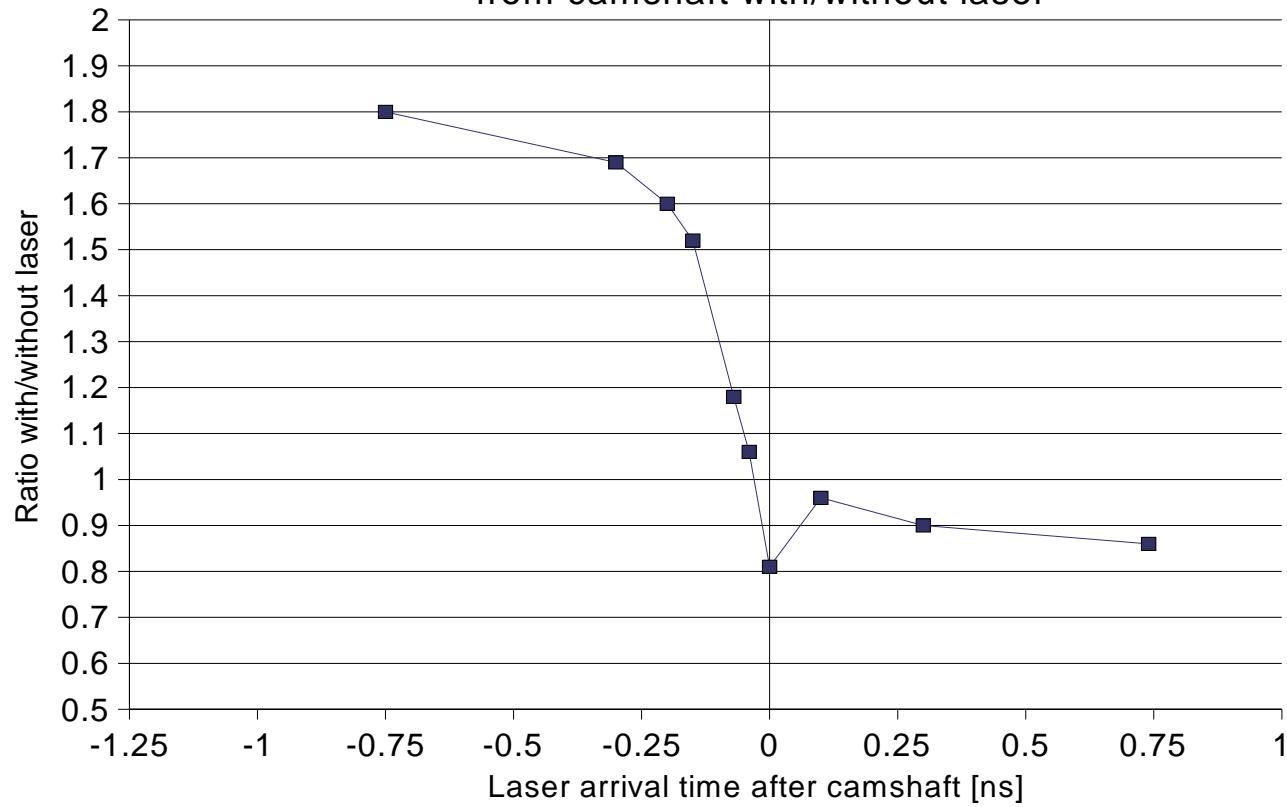
Auger electron generation in potassium



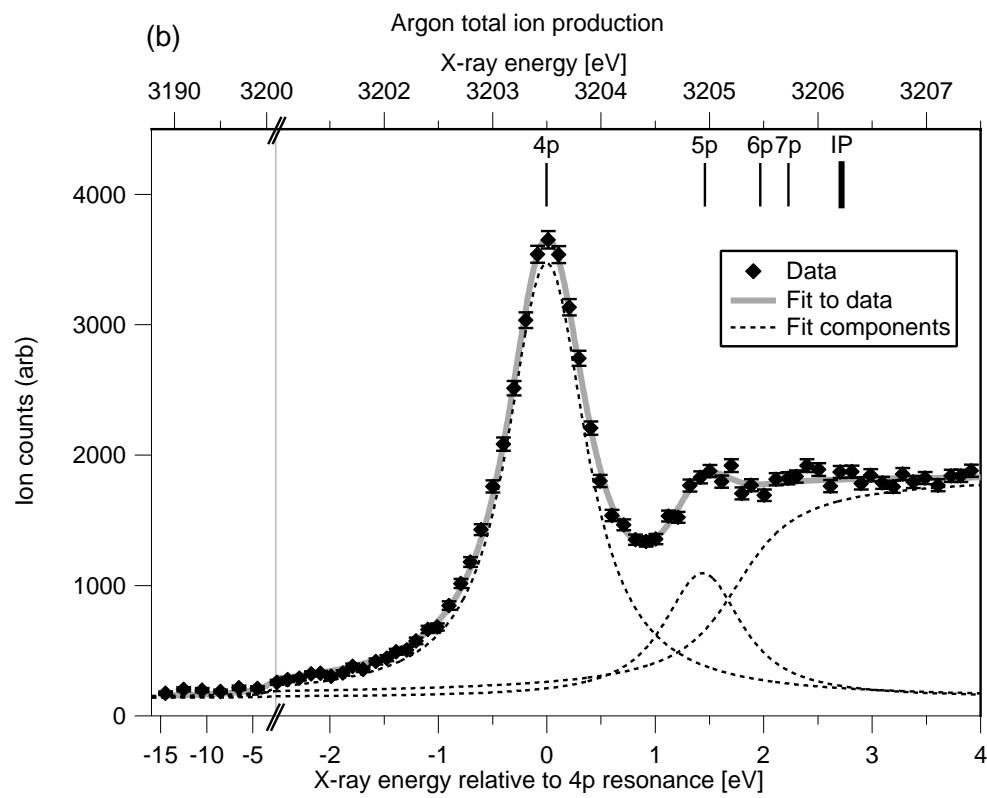




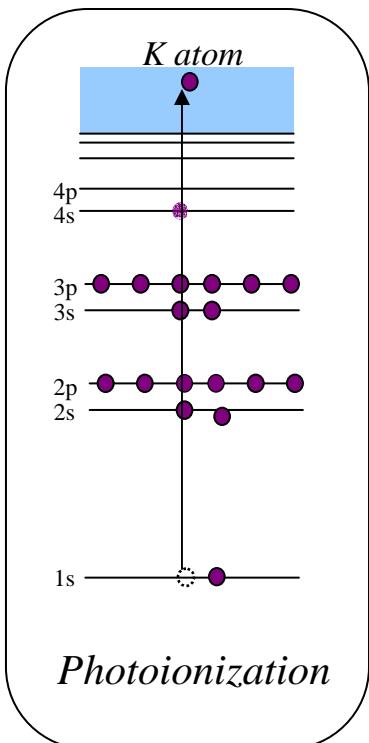
Potassium KLL Auger electron production ratio
from camshaft with/without laser



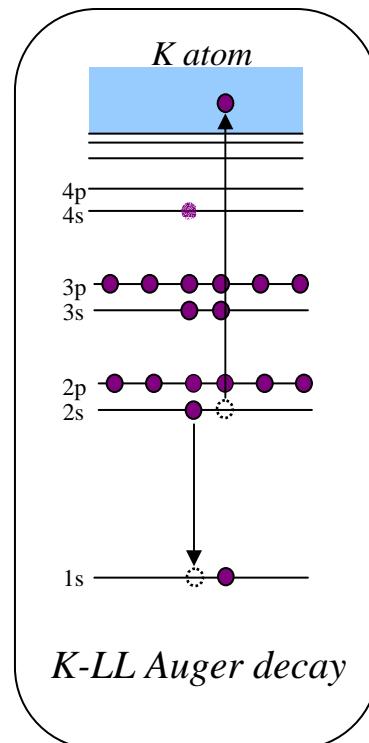
Low energy photoelectron and PCI



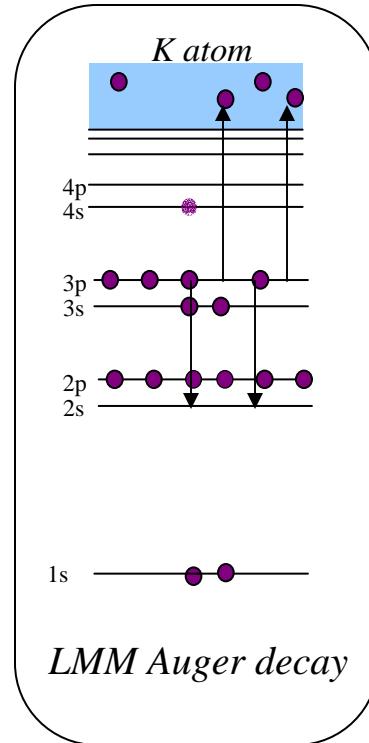
Photoionization



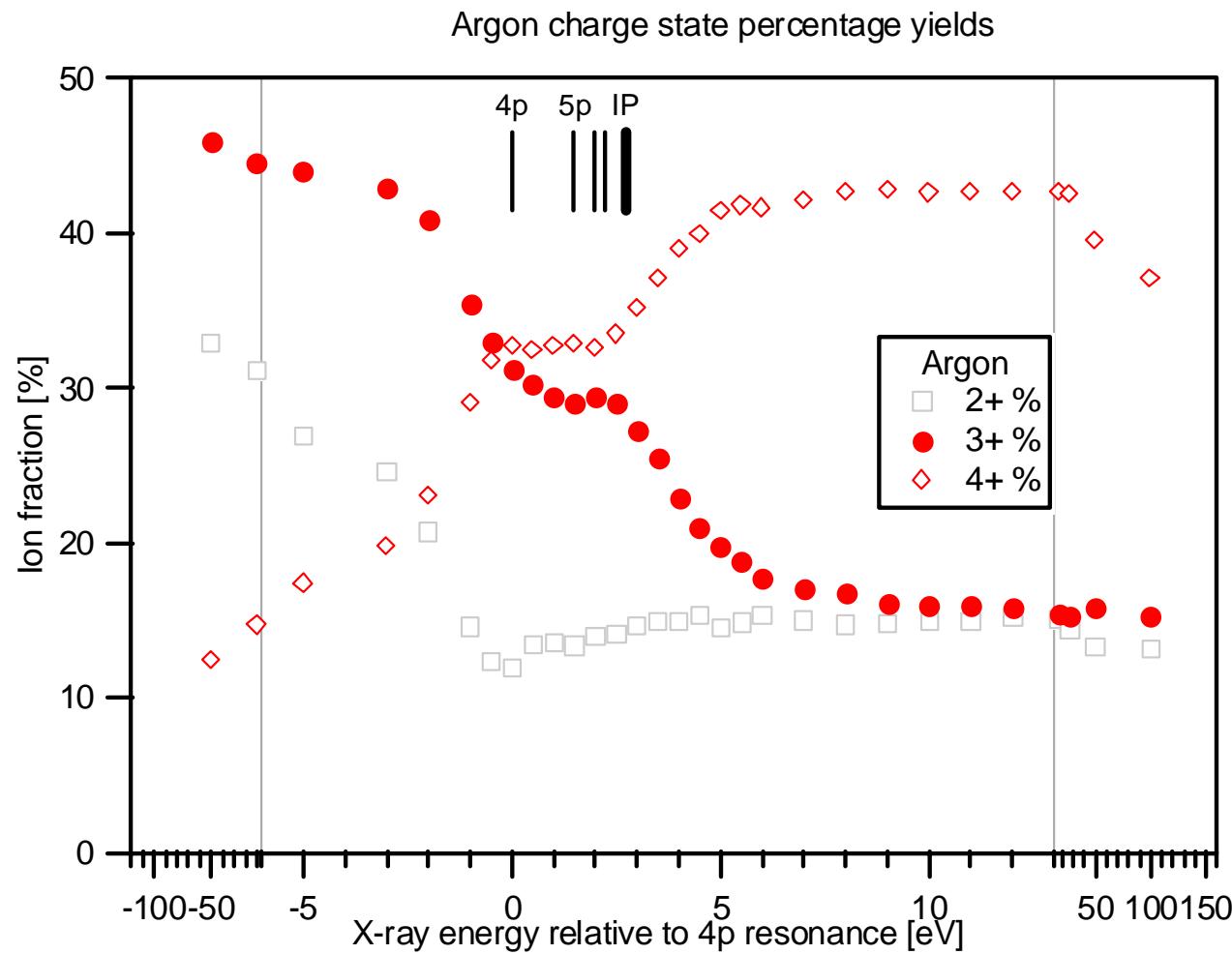
KLL Auger



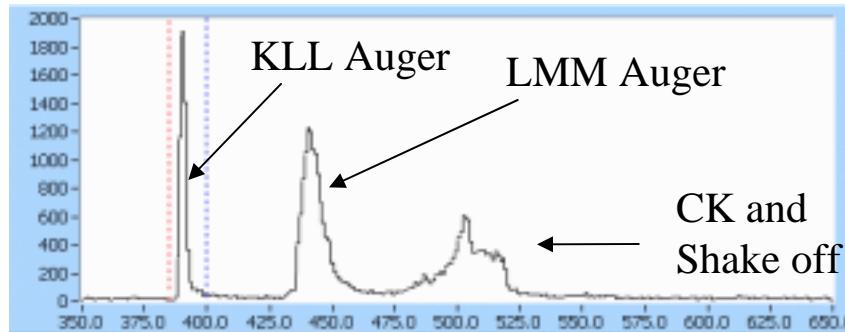
LMM Auger and multiple ionization



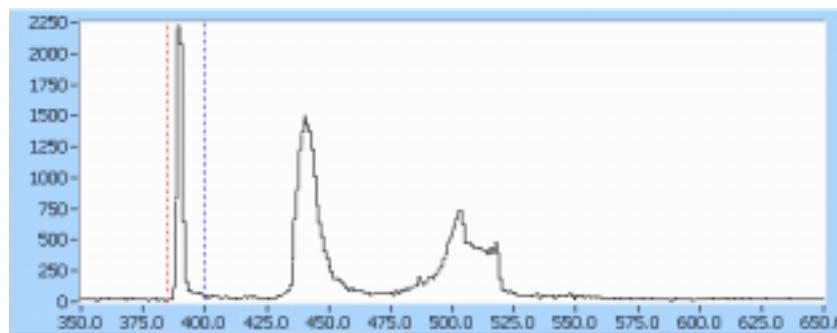
Post Collision Interaction



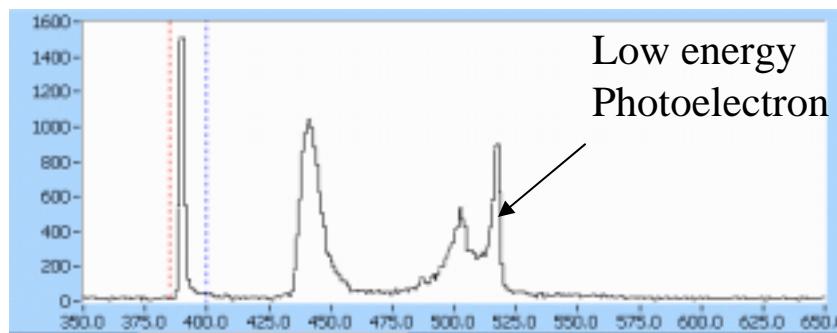
1 eV
below IP



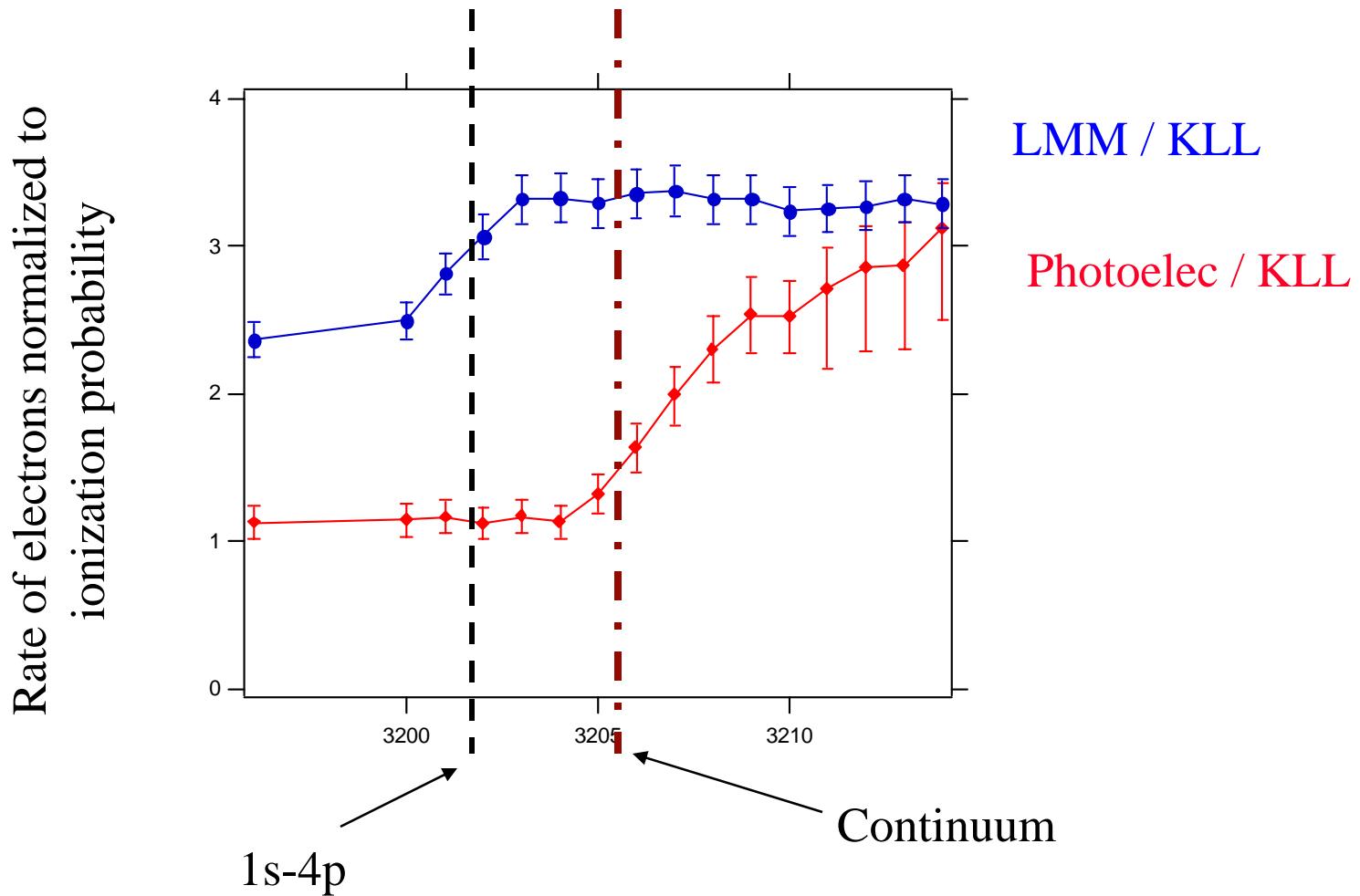
at IP



5 eV above IP



PCI and photoelectron recapture



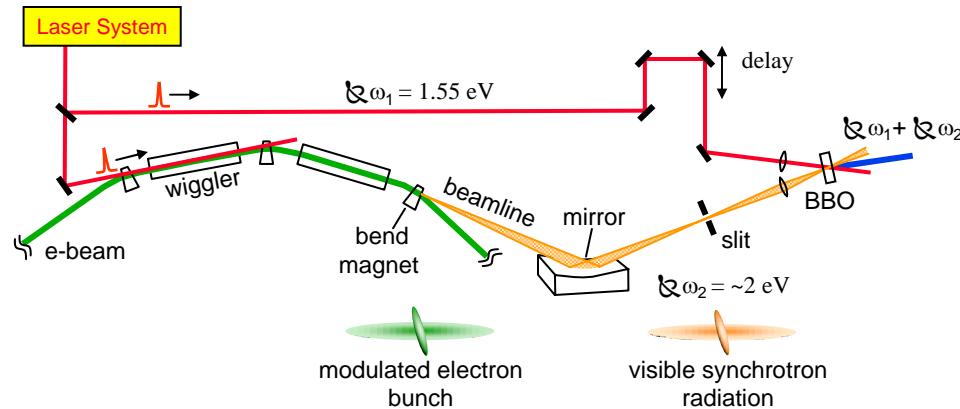
Future Plans



using femtosecond x-rays

- Study time evolution of the post collision interaction
 - short term (5.3.1)
 - Long term: **undulator beamline**
 - Ultrafast X-ray Science facility**
- Laser controlled absorption of x-rays around potassium K-edge
- Laser-assisted 1s - 4s transition in Argon
 - Non-linear strong field effects

Femtosecond Pulses of Synchrotron Radiation



Schoenlein et al., *Science*, **287**, 2237 (2000)

